On Self-Trapping in the Molecular Crystal Model in One, Two and Three Dimensions

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ON SELF-TRAPPING IN THE MOLECULAR CRYSTAL MODEL IN ONE, TWO AND THREE DIMENSIONS

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We present the outcome of a simple variational calculation for the ground-state wavefunction of the molecular crystal model (MCM) on a *lattice*. We discuss the two-site MCM and the MCM in one, two and three dimensions. For all cases we find a transition to a self-trapped state. The results seem to support our recent Monte Carlo investigations. Our variational results for the two-site MCM, including the presence of the phase transition, are in exact agreement with the rigorous solution for this model.

Recently we performed extensive Monte Carlo calculations on the molecular crystal model (MCM) [1]. The MCM is a lattice polaron with the most local action that is possible. The model has been extensively studied by Holstein and Emin [2]. According to some workers the interpretation of our Monte Carlo seems to be in conflict with well accepted ideas about the dimensionality dependence of small-polaron formation. The issue can well be summarized by stating that many of our results do not show a *qualitative* dependence on dimensionality whereas some theories would predict strong differences for the various dimensionalities. Indications for the presence of a ground-state transition in one dimension seem to surprise some workers although the presence of a ground-state transition in the *two-site* MCM, which can be considered to be the ultimate non-continuum model, can be proven rigorously [3]. The weak dependence on dimensionality becomes even more pronounced if one realizes that the results for the two-site MCM also show the same qualitative behavior as the many-site MCM in all dimensionalities. Much earlier than we did, Shore and Sander [3] already concluded that the dimensionality dependence of self-trapping is weak. The only rigorous result which could be in disagreement with the interpretation of our Monte Carlo work is the *continuum* version of the MCM in the *adiabatic* limit as discussed by Emin and Holstein [5]. Emin and Holstein have used some very elegant scaling arguments which apply to the exact wavefunction as well as to variational wavefunctions. A part of our simulations has also been done in the adiabatic regime without showing much qualitative difference with the other simulations.

In this letter we will demonstrate that the continuum model, even when supplied with a cutoff, cannot be maintained as a faithful representation of the genuine *lattice* MCM. In this paper we will give the results of a simple variational calculation of the *lattice* MCM in one, two and three dimensions in the *adiabatic* limit. Our variational theory predicts a transition in all dimen-

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sionalities. The results are in exact agreement with the rigorous two-site results and are in very good agreement with Monte Carlo work in one, two, and three dimensions [1,4]. We find that the (continuum) scaling theory breaks down for the lattice MCM. Our results cannot be obtained from a continuum scaling theory supplied with a simple cutoff. The variational calculation has been inspired by recent work by Das Sarma [6]. Das Sarma uses a gaussian variational wavefunction for a large class of continuum polarons in which he introduces lattice cutoffs to simulate the presence of a lattice. His results with respect to the presence of a (continuous or discontinuous) self-trapping transition, depend heavily on whether or not the cutoffs are introduced for some of the polarons. In our opinion the physics of the small polarons on a lattice is more subtle than the mere introduction of a cutoff in a continuum theory. This holds especially for the kinetic energy.

The hamiltonian for the MCM is given by

$$H = H_0 + H_1 + H_2 , \tag{1a}$$

$$H_0 = \frac{1}{2}\sum_{i=1}^{M} p_i^2 , \tag{1b}$$

$$H_1 = \tfrac{1}{2}\Omega^2 \sum_{i=1}^{M} x_i^2 + \lambda \sum_{i=1}^{M} x_i c_i^+ c_i , \tag{1c}$$

$$H_2 = -t \sum_{i=1}^{M} c_i^+ c_{i+1} + \text{h.c.} , \tag{1d}$$

Ω is the angular frequency of the Einstein oscillators, t is the transfer integral for the hopping motion of the electron, and λ is the electron–phonon coupling constant. The masses of the oscillators have been put equal to 1. Eqs. (1) are written for a 1D lattice, but the extension to higher dimensionalities is trivial.

Realizing that the lattice equivalent of gaussians are modified Bessel functions of the first kind, we suggest the following normalized variational wavefunction for the electron in the adiabatic limit,

$$\psi = I_0^{-d/2}(2\alpha) \sum_{n} \prod_{\mu=1}^{d} I_{n_\mu}(\alpha) c_n^+ |\text{Vac}\rangle , \tag{2}$$

where $I_n(\alpha)$ is a discretized version of the modified Bessel function of the first kind of order n and argument α which for $M \to \infty$ is identical with this Bessel function [1]. The parameter α is a variational parameter, and d stands for the dimensionality. Minimizing the expectation value of the hamiltonian with respect to $\{x_n\}$ gives

$$x_n = -(\lambda/\Omega^2)[I_0(2\alpha)]^{-d} \prod_{\mu=1}^{d} I_{n_\mu}^2(\alpha) , \tag{3}$$

The limit of x_n for $\alpha \to \infty$ is zero and in the limit $\alpha \to 0$ it is proportional to $\delta_{n,0}$. Substituting eq. (3) in the expectation value of the hamiltonian gives

$$\langle\psi|H|\psi\rangle = -2td\, I_1(2\alpha)/I_0(2\alpha) - C[I_0(2\alpha)]^{-2d} \sum_{n} \prod_{\mu=1}^{d} I_{n_\mu}^4(\alpha) , \tag{4}$$

where $C \equiv \lambda^2/2\Omega^2$. Now we have to find the minimum value of the energy as a function of the variational parameter α. It turns out that in all cases $\alpha = \infty$ is a solution of the variational procedure for all values of the effective coupling constant C. In the small-coupling regime this is the only solution for all dimensionalities. Increasing C gives rise to the appearance of a second solution (α is finite) in the variational calculation.

In one dimension this second solution has immediately a lower energy than the $\alpha = \infty$ solution. For the two-site MCM our variational calculation reproduces exactly the rigorous energy for all C including the critical value $C = 2t$ (see fig. 1). The results for the many-site MCM in one dimension are qualitatively the same as those for the two-site model (see fig. 2).

In two and three dimensions there is a certain range

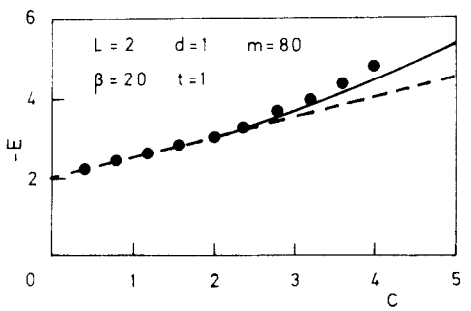

Fig. 1. Energy as a function of the scaled coupling constant C for the various MCMs in the adiabatic limit for the two-site MCM. Solid lines and dashed lines: variational results, closed circles represent data of Monte Carlo simulations. The Monte Carlo parameter m is defined in ref. [1] (L is the number of sites).

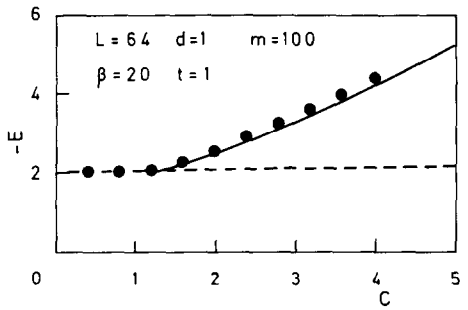

Fig. 2. Same as fig. 1 for the 1D MCM.

in C where the second solution exists but it does not have the lowest energy. After increasing C further this new solution has the lowest energy (see figs. 3 and 4).

For the purpose of comparing with our variational results we have performed additional Monte Carlo simulations. These simulations were done using the action of the MCM in the adiabatic limit. The results of these simulations have been included in figs. 1–4, and they are in good agreement with the variational approach in all dimensions. The Monte Carlo calculations have been performed at very low, but finite, temperature. If there is a phase transition in the MCM it will be a zero-temperature phase transition. When one comes in the close vicinity of the critical point by varying C the importance of a finite temperature effect becomes much more important [7]. This explains why the finite-temperature Monte Carlo results in the proximity of the critical point have a slightly higher energy than the zero-temperature variational results.

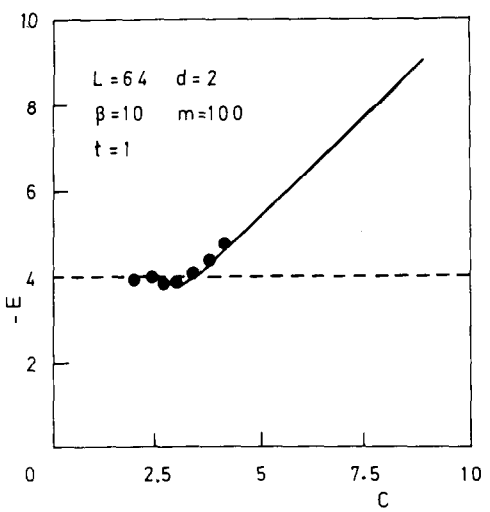

Fig. 3. Same as fig. 1 for the 2D MCM.

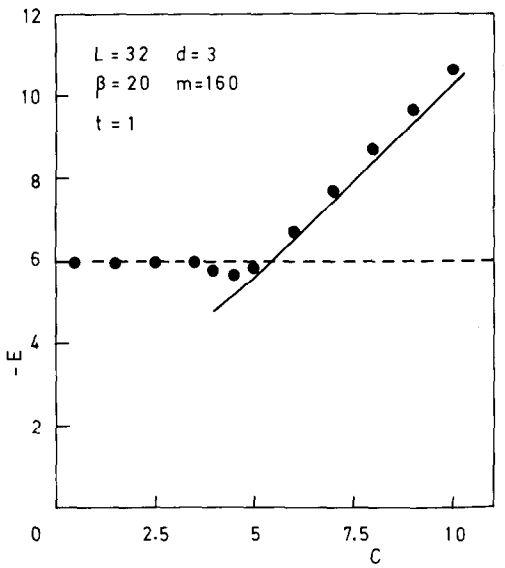

Fig. 4. Same as fig. 1 for the 3D MCM.

The interpretation of the two adiabatic solutions is quite simple. The $\alpha = \infty$ solution represents an unbound state as can be seen from eqs. (2) and (3). The finite α solutions refer to a self-trapped state as can be seen from the same equations. In all the models a critical value of C is found. This critical point is connected with a transition from an untrapped carrier to a self-trapped carrier. The results of the variational calculation for the two-site MCM reproduce the rigorous solution. In our recent Monte Carlo work we found indications for the occurrence of critical effects in the MCM at a finite value for C for all dimensionalities in agreement with the results of this variational calculation. However on the basis of a Monte Carlo calculation or a variational calculation one can never prove the existence of a transition. In addition we want to stress that conclusions drawn from the present variational calculation and from our Monte Carlo simulations refer to the MCM on a lattice only. There are many other types of polarons known and none of our conclusions should be generalized to them.

The scaling behavior of the energy of the MCM can easily be traced back. For comparison with the continuum results one should realize that α corresponds to R^2, where R is the length scale introduced by Emin

and Holstein [5]; when comparing our work with the continuum results of Das Sarma [6] one should realize that α corresponds to b^{-2} of his gaussian wavefunction. The case to compare with the continuum results is the case of large α. The expression for the potential energy goes as $\alpha^{-d/2}$ for large α in agreement with the continuum results. But the kinetic energy scales for large α as $1-4\alpha^{-1}$ fundamentally differing from the continuum result where the scaling goes as α^{-1}. It is this basic difference which is responsible in the variational calculation for the presence of a phase transition in all dimensionalities in contrast to the continuum model.

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